Sound attenuation in an unexplored frequency region: Brillouin Ultraviolet Light Scattering measurements in v-SiO_2

P. Benassi,[1] S. Caponi,[2] R. Eramo,[3] A. Fontana,[2] A. Giugni,[1] M. Nardone,[1] M. Sampoli,[4] and G. Viliani[2]
[1] *Dip. Fisica and INFM, Università dell'Aquila, I-67100 L'Aquila, Italy,*
[2] *Dip. Fisica and INFM, Università di Trento, I-38050 Povo Trento, Italy*
[3] *INFM, Dip. Fisica, and LENS, Università di Firenze, I-50019 Sesto Fiorentino, Firenze, Italy,*
[4] *INFM and Dip. di Energetica, Università di Firenze, I-50139, Firenze, Italy*
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We report Ultraviolet Brillouin light scattering experimental data on v-SiO_2 in an unexplored frequency region, performed with a newly available spectrometer up to exchanged wavevector q values of 0.075 nm^{-1}, as a function of temperature. The measured attenuation scales on visible data following a q^2 behavior and is temperature-dependent. Such temperature dependence is found in a good agreement with that measured at lower q, suggesting that its origin is mainly due to a dynamic attenuation mechanism. The comparison between the present data with those obtained by Inelastic X-ray Scattering suggests the existence of a cross-over to a different attenuation regimes.

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Our present understanding of the sound attenuation mechanisms in vitreous systems is poor compared to that of crystalline materials although this topic has attracted the interest of several researchers from both experimental and theoretical points of view [1]. In particular, the nature of vibrational dynamics of disordered systems, as derived from the study of the "low-frequency" excitations (in the hypersonic range), has been a highly debated subject in the last years [2, 3, 4, 5, 6]. Vitreous silica (v-SiO_2, or amorphous quartz) has been extensively studied as prototype of a strong glass. Nevertheless, the experimental results reported in literature had conflicting and controversial interpretations [7, 8, 9, 10, 11]. To achieve an overall characterization of this particular system is crucial to understand the vibrational dynamics of the vitreous state. It is well known that a plane wave excitation can propagate freely in a disordered structure only when the wavelength is much greater than the scale spanned by microscopic inhomogeneities. As the wavelength shortens the wave is attenuated, distorted, and scattered with increasing magnitude. As a matter of fact, the waves can even become over-damped when the wavelength approaches the interparticle spacing and the excitations become more directly affected by the microscopic disorder characteristic of the glass structure. The question about the nature of excitations far from the long wavelength limit, is unlikely to have a single answer [12, 13, 14]. Indeed different mechanisms have been suggested in order to explain the attenuation of an acoustic plane wave excitation namely: attenuation induced by topological disorder, thermally activated processes, anharmonic effects, two-level systems (TLS) processes [15]. In general, anharmonic effects are particularly relevant at relatively high temperatures while the importance of the two-level systems is confined at very low temperatures ($T < 5 \div 10\ K$). In the intermediate range $10\ K < T \ll T_g$ the TLS effects are not important and disordered induced mechanisms, thermally activated processes and anharmonic effects play the most important role in acoustic attenuation. Anyway, sound propagates through glasses and longitudinal and transverse phonons are found to be still well defined at relatively large wave-vectors (up to 5 nm^{-1}) with a linear relationship between frequency (ω) and exchanged wavevector (q) [16, 17]. Beyond this limiting q value, the effect of the topological disorder of the glassy systems causes (i) a mixing of the polarisation of the acoustic modes, which is observed for different glass forming systems both in experimental [16, 18, 19] and in molecular dynamic simulations (MDS) data [17, 20] and (ii) a possible presence of positive dispersion in the longitudinal acoustic branch [16], as theoretically predicted [21] and found in MDS [17]. Since below such q values, v-SiO_2 exhibits a substantially constant sound velocity, so the dependence of the sound attenuation on ω can be safely investigated by changing the wavevector q.

The acoustic attenuation can be measured by the full width half maximum, Γ, of the Brillouin doublet characterizing the dynamical structure factor $S(q,\omega)$. In the hypersonic region (i.e. in the GHz region investigated by the Brillouin Light Scattering technique, BLS), Γ exhibits a strong temperature dependence and suggests that the attenuation at these frequencies has to be mostly ascribed to dynamical processes [22, 23]. On the contrary, in the mesoscopic range (i.e. in the THz region investigated by the Inelastic X-ray Scattering technique, IXS) Γ has a negligible temperature dependence, supporting a non-dynamic origin of acoustic attenuation in this frequency regime [3]. In vitreous silica, the prototype of strong glasses [24], two main attenuation mechanisms have been hypothesized in the GHz and THz regions. In the first scenario [22, 25, 26] the attenuation is characterized by a crossover from a mechanism dominated by dynamical relaxation processes with a frequency dependence of $\Gamma \propto \omega^2 \propto q^2$ in the GHz range, to a mechanism dominated by strong phonon scattering,

due to presence of topological disorder in the THz region. It is hypotized that the last process should exhibit a $\Gamma \propto \omega^4 \propto q^4$ frequency dependence (much like the Rayleigh light-scattering regime by independent particles). The $\Gamma \propto \omega^4$ behaviour is able to explain the plateau in the conductivity measurements. The second scenario [3] also suggests the existence of a crossover between a low frequency (dynamical origin) attenuation mechanism and a high frequency one (nearly temperature-independent) due to topological disorder. However a $\Gamma \propto \omega^2 \propto q^2$ dependence is hypothesized for both mechanisms [3, 7, 17, 18, 27]. Such a $\Gamma \propto \omega^2 \propto q^2$ dependence valid up to high q, has also been predicted by molecular dynamic simulations performed on realistic v-SiO_2 [28], on harmonic glasses [29] as well as on hard spheres systems [21] and disordered linear chains [30]; it is also supported by recent theoretical free-energy landscape studies [31, 32].

BLS and IXS technique do not cover the entire frequency (and q) range from GHz to THz and investigations within the frequency gap which separates these techniques could be useful to discriminate between the different hypotheses. As a matter of fact, between the GHz and THz region, the sound attenuation has already been measured by the picosecond optical technique (POT) [33] and no dependence on temperature of Γ has been found in the range $80 \div 300\ K$. Unfortunately, these difficult measurements are affected by large systematic errors and random uncertainties. In the region where there is an overlapping with the BLS attenuation data, the POT values of Γ are always more than a factor 2 larger. As a consequence, even if the POT data can give a qualitatively insight of the q behaviour, they are of little help in understanding the different behaviour of the temperature dependence of Γ found by BLS and IXS techniques. In order to shed some light on the underlying mechanisms an accurate investigation of the attenuation in the intermediate q-region. This is now possible thanks to the development of a new spectroscopic apparatus operating in the ultraviolet region [34]. Here we report the investigation of ultraviolet Brillouin light scattering (BUVS) on v-SiO_2 (spectrosil) performed at different temperatures. We find that, in the BUVS regime, the measured line-width Γ appears to be temperature dependent with a q^2 behaviour. Our findings are in agreement with the existence of two main attenuation mechanisms, with a relative weight which is dependent on the phonon wavelength. They are also compatible with the existence of a crossover between a prevalent dynamic and a prevalent static sound attenuation mechanism and give some hints for a possible explanation of the Γ excess found by extrapolating the q^2 behavior of IXS data down to BLS.

The experimental apparatus used for the measurements with ultraviolet excitation consists of a new-built spectrometer with high resolution, contrast, and luminosity, called HIRESUV [34]. The instrument is based on a double grating 4 meter focal length monochromator specifically designed for Brillouin spectroscopy with both visible (532 nm) and ultraviolet (266 nm) excitation. The high luminosity is achieved with the two large echelle grating (400x208 mm with 31.6 $grooves/mm$). The size of the gratings and mirrors yield an instrumental $F-$number of 1 : 40 in the vertical plane (where dispersion occours) and of 1 : 20 in the horizontal plane. In the UV range, HIRESUV works at the 230-th order and reaches a resolution of about 0.6 GHz. To overcome air turbulence effects and small changes of refractive index and further to warrant a good thermal conductivity, the entire apparatus, except the sample compartment, is inside a vacuum chamber filled with helium at atmospheric pressure. All the optical items (mirror, slits, and light collecting lenses) are positioned and aligned by means of computer controlled microstep movements. The photocounts of a low noise phototube are recorded at fixed wavelength for subsequent elaboration. The UV radiation is generated by a commercial system based on the second-harmonic generation of a visible laser source. Other details of HIRESUV will be published elsewhere [34]. A typical spectrum obtained using HIRESUV at room temperature is reported in fig. 1 (the measured resolution in this case was set to about 0.8 GHz) where the high contrast and high resolution reached by the experimental apparatus can be appreciated. The BUVS signal (open circles), proportional to the dynamic structure factor, $S(q,\omega)$, convoluted with the instrumental resolution function, $R(\omega)$, is reported in a log scale together with a fit function (full line). To get quantitative information on characteristic frequency and on the attenuation Γ of the excitations, the data have been fitted by the convolution of the experimentally determined $R(\omega)$ with the sum of an elastic and an inelastic contribution; the former, is represented by a δ-function while the latter has been described by a DHO model.

As an example of the temperature behaviour of the Brillouin peak, the Anti-Stokes parts of the spectrum for two selected temperatures (open circles) together with their resolution curves (dots connected by lines) and the best fit (full lines) are reported in fig. 2. Directly from the rough BUVS data one can visualize the temperature changes of the width, indicating that, in this frequency range, at least part of the sound attenuation has a dynamical origin. In the inset of fig. 1, the HWHM of the Brillouin peak, once scaled by q^2, as a function of temperature is reported; the temperature behaviour seems to be in good agreement with the one obtained by BLS measurements [22]. The values of $\Gamma(q)$ measured at room temperature resulting from the fits of BUVS (dots) and BLS (full diamond, circle [22] and open triangle [35]) data are reported in fig. 3. The spectra collected at 90°-scattering angle were measured with an horizontal collection angle of $\delta\phi_H = 1.7\ mrad$. In this configuration the exchange wavevector dispersion arising from the finite acceptance angle, $\delta q/q = \delta\phi_H/2$ gives a negligible contribute to the the mode linewidth. The dashed lines represents the extracted q^2 behaviour. In the same figure we have reported the POT data (asterisks [33]) and IXS

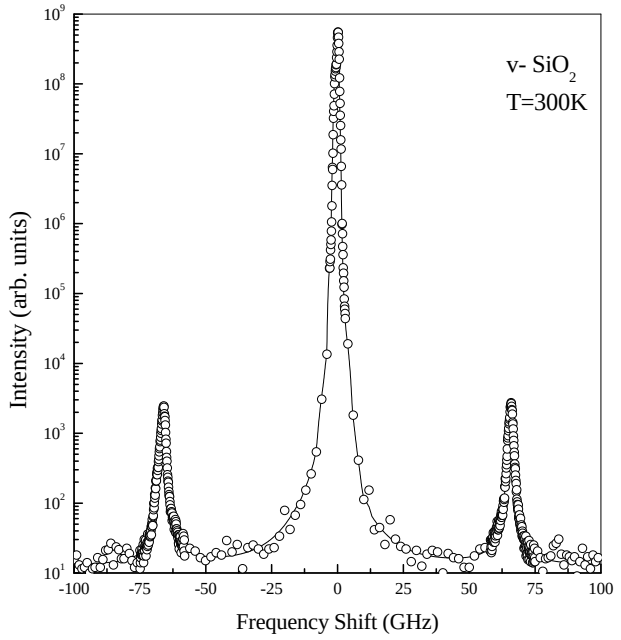

FIG. 1: Typical Brillouin ultraviolet Light scattering spectrum on v-SiO_2 at $T = 300\ K$ (180° scattering geometry and the excitation wavelength was 266 nm). The experimental data (circles) are reported together with the best fit (full line) see text. The excitation wavelength was 266 nm, with a power $\approx 100\ mW$.

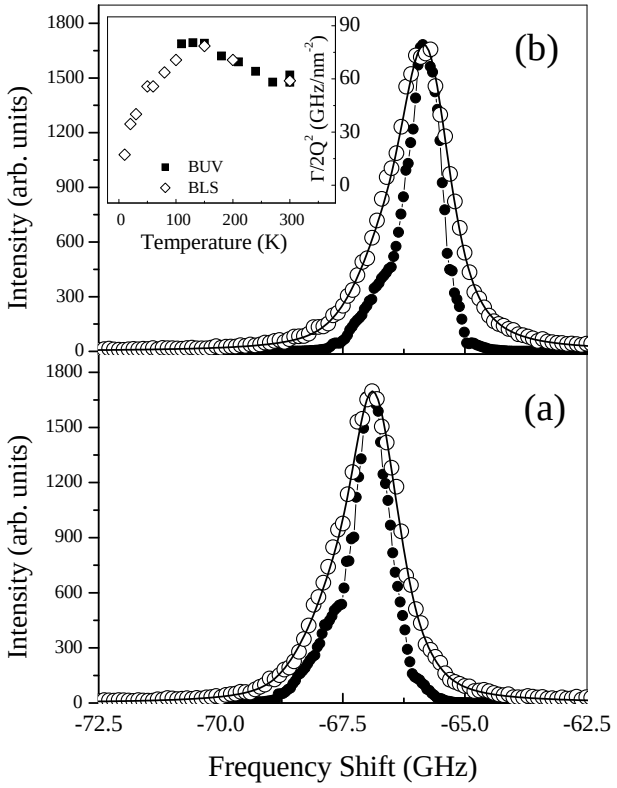

FIG. 2: Anti-Stokes part of BUVS spectra (circles) at two significant temperatures: a) at 270 K, b) at 150 K. The resolutions (full lines and dots) and the best fit (full lines) are also reported. In the inset is shown the temperature behaviour of the $\Gamma/2q^2$ for the BLS (open diamonds)and BUVS (full squares) data.

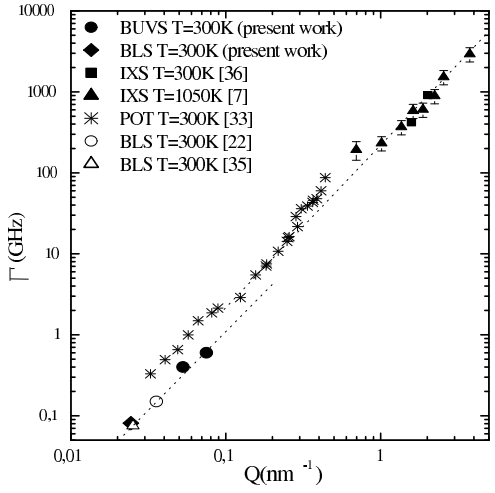

FIG. 3: Log-log plot of Brillouin widths as a function of exchanged wavevector q obtained at room temperature by BLS (full diamond, open triangle [35], open circle [22]), BUVS (full circle) in 90° and 180° scattering configurations, and IXS (triangles $T = 1050\ K$, squares $T = 300\ K$). Asterisks represent the results obtained by POT technique [33]. The dashed lines represent the q^2 law for the respective data.

data at $T = 1050\ K$ (full triangles [7]) and at $T = 300\ K$ (full squares [36]). The new BUVS data are along the same BLS trend while BLS and BUVS data are always more than a factor 2 below the POT data. In our opinion, this confirms that the POT data can only give a qualitative information. At high q, inside the experimental error, $\Gamma(q)$ obtained by IXS do not show any noticeable temperature dependence in the $300 \div 1500\ K$ investigated range [3] and the measured values are in good agreement with the ones calculated by MDS for the extreme $T = 0$ case [28]. The $\Gamma(q)$ is well fitted to a q^2 law in the $1 \div 5\ nm^{-1}$ range confirming, again, the general trend found in the simulations [28]. Within the experimental uncertainties, the temperature independence of Γ indicates that most of the attenuation has a non-dynamical origin and is ascribable to the presence of topological disorder [3]. It is worth to notice that a q^2 law is not able to reproduce accurately both high-q IXS and low-q BLS and BUVS data at room temperature. Therefore $\Gamma(q)$ must have a more complex behaviour than a q^2 dependence: between $1\ nm^{-1}$ down to $0.1\ nm^{-1}$ it should decrease more steeply in order to join the q^2 behaviour at low-q values. Temptatively we will write the attenuation Γ as a sum of two contributions: $\Gamma(q,T) = \Gamma_s(q) + \Gamma_d(q,T)$, where $\Gamma_s(q)$ is the structural part dependent on the topological disorder at the nanometer scale while $\Gamma_d(q,T)$ is due to dynamical temperature dependent processes effective at the micrometer scale. The $\Gamma_s(q)$ part is independent on temperature since the topological structure

at molecular level is insensitive to temperature changes, as confirmed by the measurements of the static structure factor in many glasses. It is the dominant contribution at higher q values. On the contrary, when the wavelength is much larger (in BLS range) and the system is viewed as a continuum, the disorder produced by dynamical processes becomes the major responsible for the attenuation and the contribution related to the structural disorder, $\Gamma_s(q)$, will be lower than the Γ minimum value reached around 5 K (about two order of magnitude smaller than the 300 K value) [22]. The interplay between the two mechanisms, in the intermediate frequency region, results in a cross-over from a temperature dependent to a temperature independent sound attenuation, depending on which one of the to processes is the dominating mechanisms. The BUVS measurements, performed as a function of temperature, indicate a dynamic contribution up to exchange wavevector values of $q \approx 0.075\ nm^{-1}$. In conclusion in this work, we have reported the measure of the Brillouin spectra performed employing UV excitation on v-SiO_2, using the new HIRESUV spectrometer. The experimental data obtained in the, up to now, unexplored frequency region give indications that in vitreous silica the dominant sound attenuation mechanism has a distinct origin in the different q regions spanned by the IXS or BUVS/BLS techniques. Notwithstanding the complex behaviour of $\Gamma(q,T)$, which appear to be far from a simple square law, the data do not support the first hypothesis on the origin of sound attenuation [22, 25, 26]. Indeed, in the high frequency or exchange wavevector range, the attenuation is never found to be growing with a fourth power law and moreover, even if in the unexplored q range (between 0.1 nm^{-1} and 1 nm^{-1}) a steeper than q^2 dependence has to be expected, at high q values the q^2 behaviour is again recovered. Concerning the second hypothesis, other measurements in the unexplored frequency region are necessary to verify the predicted existence of the cross-over region. Moreover we want to stress that, in the framework of second hypothesis, the excited sound plane wave inside the glass in the THz range is far from a plane wave; on the contrary it is formed by a large spectral combination of eigenvectors, and the loss of phase relationship between them is the major cause of the plane wave attenuation. This attenuation is not effective for the thermal conductivity which is related to the decay of a given eigenvector amplitude and not to a phase change between different eigenvector components. This could explain qualitatively why the thermal conductivity behaves so differently from sound attenuation and further earns high marks for the second hypothesis.

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